

Personal and Outdoor Nitrogen Dioxide Concentrations in Relation to Degree of Urbanization and Traffic Density

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To assess differences in exposure to air pollution from traffic in relation to degree of urbanization and traffic density, we measured personal and home outdoor nitrogen dioxide (NO₂) concentrations for 241 children from six different primary schools in the Netherlands. Three schools were situated in areas with varying degrees of urbanization (very urban, fairly urban, and nonurban) and three other schools were located near highways with varying traffic density (very busy, fairly busy, and not busy). Weekly averaged measurements were conducted during four different seasons. Simultaneously, indoor and outdoor measurements were conducted at the schools. Personal and outdoor NO₂ concentrations differed significantly among children attending schools in areas with different degrees of urbanization and among children attending schools in areas close to highways with different traffic densities. For the children living near highways, personal and outdoor NO₂ concentrations also significantly decreased with increasing distance of the home address to the highway. Differences in personal exposures between children from the different schools remained present and significant after adjusting for indoor sources of NO₂. This study has shown that personal and outdoor NO₂ concentrations are influenced significantly by the degree of urbanization of the city district and by the traffic density of and distance to a nearby highway. Because NO₂ can be considered a marker for air pollution from traffic, the more easily measured variables degree of urbanization, traffic density, and distance to a nearby highway can all be used to estimate exposure to traffic-related air pollution. **Key words:** distance, exposure, highways, nitrogen dioxide, traffic, urbanization. — *Environ Health Perspect* 109(suppl 3):411–417 (2001). <http://ehpnet1.niehs.nih.gov/docs/2001/suppl-3/411-417rijnders/abstract.html>

During the last decade, air pollution in relation to respiratory health has again become an important issue. In addition to indoor sources, automobile traffic has been recognized as a major source of air pollution exposure. Traffic emissions consist of volatile hydrocarbons, airborne particles, nitrogen oxides, and carbon monoxide. Several recent studies suggest an association between air pollution from traffic and adverse effects on respiratory health (1–6). In many of these studies, crude measures of exposure to traffic-related air pollution, such as traffic density on the street of residence and/or distance of the home address to busy roads, are used (1,2). Some other studies have used air pollution levels measured at a central ambient site or in the schools of the children (3–5). Few studies have incorporated personal measurements of traffic-related air pollution, either as a direct measure of exposure or as a validation of the exposure measures used (6,7).

Several studies show that ambient air pollution from traffic is higher in urban areas than in rural or nonurban areas (8–10). For example, in the PEACE study, a multicenter study of acute pollution effects on asthmatic children in Europe, the median urban/rural ratio pooled over all 14 locations was 1.8 for nitrogen dioxide (NO₂) and 1.4 for black smoke (8). Although none of these studies have related air pollution concentrations to some

measure of the actual degree of urbanization, it can be expected that with increasing degree of urbanization, for example, expressed as the number of addresses per unit area, exposure to traffic-related air pollution will increase as well.

Studies have shown also that air pollution from traffic is higher along busy roads compared to background locations (11–13). Air pollution from traffic in city districts near highways is related to the traffic density of the highway, distance of the measuring site to the highway, and the percentage of time that the measuring site was downwind of the highway (14). The range in concentrations between the city districts located along highways with different traffic densities was larger than the variation with distance from the highway within city districts. It was therefore suggested that exposure to traffic-related air pollution varies less among subjects living within the same city district than among subjects living along highways with different traffic densities.

This study was designed to test two (null) hypotheses: *a*) there is no difference in exposure to NO₂ as a marker of traffic-related air pollution among subjects living in areas with a different degree of urbanization; and *b*) there is no difference in exposure to NO₂ as a marker of traffic-related air pollution among subjects living close to highways with different traffic intensities.

Materials and Methods

We used NO₂ as a marker for traffic-related air pollution (3,6). Personal exposure to NO₂ was measured in children from six different schools. Three schools were situated in city districts with varying degrees of urbanization, with no busy streets within 300 m of the schools. The other three schools were within 400 m of highways with varying traffic densities. In addition to personal measurements of the children, parents were asked to perform outdoor NO₂ measurements at the back side of their homes. Weekly averaged measurements were conducted during four different seasons.

Study Locations

For the first part, three schools were chosen located in areas with various degrees of urbanization. The degree of urbanization, developed by the Dutch Central Bureau of Statistics, is based on the average address density per unit area. It classifies all Dutch municipalities in 5 degrees of urbanization, with 1 being the most-populated level and 5 the least-populated level, so that each group has approximately the same number of inhabitants. Three schools from municipalities with a degree of urbanization of 1 (very urban), 3 (fairly urban), and 5 (nonurban) were selected from all schools in the center of the Netherlands (Utrecht province). Schools were selected on the basis of two criteria: *a*) the degree of urbanization of the postal code area of the school was the same as for the whole municipality; and *b*) the schools were more than 300 m from busy roads. The urban densities of the postal code areas of the selected schools were 3,792; 1,481; and 195 addresses per square kilometer for degree of urbanization of 1, 3, and 5, respectively. The years of construction of the school buildings were 1937, 1985, and 1967 for degree of urbanization of 1, 3, and 5, respectively.

For the second part of the study, 3 schools were chosen from 24 schools participating in

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a study on health effects of exposure to traffic-related air pollution of children attending schools within 400 m of highways (15). In the selection of the schools, goals were a maximum variation in total traffic density and a minimum variation in the geographic locations of the schools with respect to the highways. The latter criterion was used to reduce differences in the percentages of time that the schools would be downwind during the measurements. The selected schools were situated in the southwest of the Netherlands. The schools were classified as very busy (169,637 vehicles/24 hr), fairly busy (126,115 vehicles/24 hr) and not busy (45,129 vehicles/24 hr) according to the total traffic density of the nearby highway. Characteristics of the selected schools are listed in Table 1. The school along the highway with the lowest traffic density was in an area with a lower degree of urbanization and was closer to the highway than the other two schools. The geographic location of this school with respect to the highway also deviated approximately 29° from the geographic location of the two other schools. The school building along the very busy highway was older (year of construction, 1929) than the other two school buildings (years of construction, 1973 and 1979 for the school along the fairly busy and not busy highway, respectively).

Sampling Strategy

Children between 6 and 12 years of age were asked to participate in the study. All children of a class were asked to participate by handing out information letters and informed consent forms for their parents. Two to four classes per school were approached, depending on the number of children per class. A total of 397 children were asked to participate in the study. The study started in the autumn with 222 (56%) children. In the winter 19 additional children volunteered to participate. One new child started in the spring. During the course of the study, 10 children dropped out.

Personal NO₂ exposure of the children and outdoor NO₂ at the back side of their homes were measured during 1 week in four different seasons. Concentrations of indoor NO₂ in the classrooms and outdoor NO₂ at the back side of the schools were measured in the same week. During autumn, winter, and spring, measurements were conducted for all

six schools simultaneously. For logistic reasons it was not possible to conduct all measurements in the same week in the summer. In the summer, therefore, the measurements of the urbanization schools and traffic schools were conducted in 2 separate weeks. The school along the highway with the highest traffic density did not participate during the summer season.

Sampling sites of indoor measurements at school were located in the classrooms of the participating children, away from windows and doors. Outdoor measurements at the schools were conducted at the back side of the school at approximately 1.5–2 m above the ground. Children received verbal instructions from one of the investigators at the school on how to take care of their samplers. Passive sampling tubes were attached to a badge and worn between breast and head. Outdoor samplers were given to the children, with written instructions for the parents. With the aid of photographs and examples, these written instructions explained the use of the personal exposure badges of their children and the measurement of outdoor NO₂ at their homes. Personal exposure sampling was initiated and stopped at school. In small groups of five, children were asked to uncup tubes and after 1 week, to recap their tubes under supervision. The supervising researcher registered date and time. Parents were instructed to attach the outdoor tubes to the back side of their homes using a specially designed device and to write down on a form the time of uncapping and recapping. The sampled outdoor tubes were handed in by the children on the same day that the personal exposure samples were collected. Tubes handed in after the collection day were sent back by mail by the teacher, using a preaddressed envelope. Collected tubes were sealed and stored in a refrigerator until analysis (within 3 months). To motivate the children, teachers were encouraged to wear NO₂ samplers as well. After each measurement, children were given a small present. After winter the children were given a presentation on the progress of the study to further motivate them.

Sampling Methods

NO₂ was measured using diffusion tubes described by Palmes et al. (16). These tubes consisted of a transparent cylinder closed at

one end with a red cap containing a metal grid coated with triethanolamine. All measurements were conducted in duplicate, and the average was used in the statistical analyses. A total of 902 pairs of personal sampling tubes and 842 pairs of outdoor sampling tubes were handed out. The number of outdoor samples was lower than the number of personal tubes because if more than one child in the same household participated, tubes for outdoor sampling were given only to the oldest child. As a result, 17 children from 16 different households did not receive any outdoor tubes. A total of 839 (93%)-sampled personal duplicates and 750 (89%)-sampled outdoor duplicates were collected and analyzed. Five personal and 28 outdoor concentrations could not be calculated because of missing information in the sampling times. Another 7 outdoor concentrations were excluded because the measurement time was less than 4 days. In addition, 43 personal duplicates and 7 outdoor duplicates were excluded because of bad reproducibility (coefficient of variation [CV] > 25%). The mean CV of the remaining duplicates was 6.7% (SD 5.8; median 5.2%) for personal measurements and 6.3% (SD 5.5; median 4.7%) for outdoor measurements. In total, 791 (88% of distributed) personal and 708 (84% of distributed) outdoor concentrations were obtained. These also include measurements of children who participated in fewer than four seasons.

Exposure Variables

Information on potential indoor sources of NO₂ in the homes of the children and other characteristics was assessed using a self-administered questionnaire distributed at the beginning of the project.

For children from the three schools near a highway, the distance of the home from the highway, defined as the distance between the center of the postal code area of the home address to the highway, was calculated using a geographic information system (GIS). In addition, wind direction was used to evaluate the percentage of time the schools were downwind of the highway during the measurement periods. Data on wind direction per hour were obtained from Rotterdam Airport Zestienhoven of the Royal Dutch Meteorological Institute. The percentage of time that a school had been downwind of the highway was calculated by determining a 120° sector surrounding the perpendicular line connecting the school to the highway (14).

Data Analysis

First, the distribution of personal and outdoor NO₂ concentrations was calculated for each school and season separately. Differences among the schools were tested using a *t*-test. Because of the known strong

Table 1. Characteristics of the three schools situated close to a highway.

Category	Highway	Total traffic ^a	Car traffic ^b	Truck traffic ^c	Orientation ^d	Distance from highway (m)	Degree of urbanization	
							Level	Addresses/km ²
Very busy	A20	169,637	149,450	20,187	168°	349	1	2,950
Fairly busy	A20	126,115	105,810	20,305	169°	335	1	2,689
Not busy	A58	45,129	39,939	5,190	197°	62	3	1,252

^aNumber of vehicles per 24 hr. ^bNumber of vehicles < 5.1 m length per 24 hr. ^cNumber of vehicles > 5.1 m length per 24 hr. ^dGeographic location with respect to the highway.

influence of unvented gas water heaters on indoor NO₂ concentrations in the Netherlands (17,18), children whose parents reported that they had such a water heater were excluded from the analysis.

Combined analysis of data from all four seasons was performed using a multiple regression model in which three dummy variables for each season were included. The SAS procedure "Proc Mixed" (SAS Institute, Cary, North Carolina, USA) was used to adjust regression results for correlations between repeated measurements of the same child. A random intercept model was used. The all-seasons combined effect of degree of urbanization or traffic density on personal and outdoor NO₂ was calculated using both the categorical urbanization/traffic density levels (very, fairly, non) as the continuous urbanization/traffic density variables (addresses per square kilometer and vehicles per 24 hr for urbanization and traffic density, respectively).

For both groups of three schools, combined season analyses for personal exposure were also calculated after adjusting for the following potential indoor sources of NO₂: unvented gas water heater, vented gas water heater, gas cooker, parental smoking, and a gas space heater in the living room. For the traffic density schools the distance between the home address and the highway was added to the model. We expected an exponential decay of the concentrations with increasing distance from the road (14), so we used the logarithm of the distance. Furthermore, we evaluated any significant differences in the

personal/outdoor ratios between the schools that, if present, could point to differences in personal exposures between the schools caused by factors other than the outdoor NO₂ concentrations or the indoor sources mentioned previously.

Results

Population

Characteristics of the participating children are shown in Table 2. The number of participants was smallest for the schools in the area with the highest degree of urbanization and along the busiest highway. Potential indoor sources were generally present more in the homes of the children from the highest exposure category, especially for the urbanization schools. In total, 14 children lived in a home with an unvented gas water heater ("geiser"). In most households, gas was used for cooking.

NO₂ Concentrations and Degree of Urbanization

Results of the classroom and outdoor NO₂ measurements at the schools in areas with varying degrees of urbanization are presented in Table 3. Except for the classroom concentrations in autumn, classroom concentrations and outdoor concentrations at the back side of the schools were in all seasons highest in the area with the highest degree of urbanization and lowest in the area with the lowest degree of urbanization. The difference among average classroom concentrations in the very urban and nonurban school (12.2 µg/m³) was

somewhat larger than the difference among the average outdoor concentrations (11.4 µg/m³). This indicates that the indoor/outdoor ratio (I/O) was larger for the very urban school (yearly average I/O = 0.6) compared to the other two urbanization schools (yearly average I/O = 0.4). As there are no important indoor sources of NO₂ in these classrooms, this is possibly the result of higher ventilation of the very urban school in the very urban area. This is supported by the fact that this school building was much older (pre-World War II) than the other two school buildings.

The distributions of personal and home outdoor NO₂ concentrations in areas with varying degrees of urbanization are presented in Table 4. In all seasons, both personal and outdoor NO₂ concentrations were highest in the area with the highest degree of urbanization and lowest in the area with the lowest degree of urbanization. The difference between outdoor concentrations in the very urban and nonurban areas was significant in all seasons except autumn, which may be due to the low number of outdoor concentrations in the very urban area (*n* = 9). When comparing the very urban with the fairly urban area, we found significant differences (*p* < 0.05) in autumn, winter, and spring for personal exposures, and in winter, spring, and summer for outdoor concentrations.

When the data from all seasons were combined, the estimated difference between the area with the highest degree of urbanization and the area with the lowest degree of urbanization was 14.6 µg/m³ (standard error of the mean [SE] 1.7) for personal exposures

Table 2. Characteristics of the participating children.

Characteristic	Urbanization schools			Traffic schools		
	Very urban (<i>n</i> = 33) 3,792 addresses/km ²	Fairly urban (<i>n</i> = 56) 1,481 addresses/km ²	Nonurban (<i>n</i> = 43) 195 addresses/km ²	Very busy (<i>n</i> = 27) 169,637 vehicles/24 hr	Fairly busy (<i>n</i> = 39) 126,115 vehicles/24 hr	Nonbusy (<i>n</i> = 44) 45,129 vehicles/24 hr
	<i>n</i> (%)	<i>n</i> (%)	<i>n</i> (%)	<i>n</i> (%)	<i>n</i> (%)	<i>n</i> (%)
Girls	19 (58)	32 (57)	24 (56)	12 (48)	20 (51)	22 (50)
Unvented gas water heater	5 (15)	0 (0)	0 (0)	5 (20)	2 (5)	2 (5)
Vented gas water heater	8 (24)	1 (2)	2 (5)	3 (12)	0 (0)	6 (14)
Cooking with gas	33 (100)	48 (86)	29 (67)	24 (100)	31 (82)	34 (79)
Parental smoking	22 (82)	24 (43)	22 (51)	17 (68)	14 (36)	28 (64)
Gas space heater in living room	3 (9)	5 (9)	15 (35)	2 (8)	3 (8)	8 (18)
Dutch origin	10 (33)	52 (93)	43 (100)	21 (84)	33 (85)	36 (82)
Low-education ^a mother	15 (58)	3 (6)	2 (5)	8 (36)	2 (5)	8 (18)
Low-education ^a father	16 (57)	3 (6)	5 (13)	2 (10)	1 (3)	6 (14)
Home within 400 m of the highway				16 (64)	20 (51)	32 (74)
Home within 1,000 m of the highway				21 (84)	32 (82)	43 (100)

^aPrimary school only.

Table 3. Mean classroom and outdoor school NO₂ concentrations at schools in areas with varying degrees of urbanization.

Period (dd/mm/yy)	Classroom mean ^a			Outdoors school		
	Very urban 3,792 addresses/km ²	Fairly urban 1,481 addresses/km ²	Nonurban 195 addresses/km ²	Very urban 3,792 addresses/km ²	Fairly urban 1,481 addresses/24 km ²	Nonurban 195 addresses/km ²
Autumn (24/11/97–01/12/97)	20.9	8.0	9.3	31.6	27.6	26.7
Winter (10/02/98–17/02/98)	29.4	16.0	12.4	59.0	41.6	33.2
Spring (17/04/98–24/04/98)	22.9	13.4	12.5	32.4	29.2	26.8
Summer (19/06/98–26/06/98)	19.0	12.4	9.6	24.8	20.5	15.7
Average	23.1	12.5	10.9	37.0	29.7	25.6

^aMean of 2–4 classrooms.

and $11.0 \mu\text{g}/\text{m}^3$ (SE 0.9) for outdoor concentrations. The estimated differences between the area with the intermediate degree of urbanization and the area with the lowest degree of urbanization were $3.9 \mu\text{g}/\text{m}^3$ (SE 1.3) for personal exposures and $5.0 \mu\text{g}/\text{m}^3$ (SE 0.6) for outdoor concentrations. Table 4 shows that for the school in the very urban area the range in personal NO_2 concentration was large in both winter and summer, which in both seasons is caused by an outlying high concentration from the same child (who lived in a home with a vented gas water heater). Removing these outliers somewhat decreased the estimated difference in personal exposures between the very urban and nonurban area (from 14.6 to $12.3 \mu\text{g}/\text{m}^3$). When the degree of urbanization of the home address in addresses per square kilometer instead of the categoric urbanization level of the school was included in the model, an increase in NO_2 concentrations of $3.4 \mu\text{g}/\text{m}^3$ per 1,000 addresses per km^2 was estimated for both personal and outdoor concentrations (SE 0.4 and 0.3, respectively).

NO_2 Concentrations and Traffic Density

Table 5 shows the percentages downwind, classroom concentrations, and outdoor

concentrations during the measurement periods. On average, the percentage of time that the school was downwind of the highway during the measurements was 15% higher for the school with the lowest traffic density compared to the other two schools. Outdoor NO_2 concentrations at the back side of the schools were, on average, highest for the school along the very busy highway and lowest for the school along the nonbusy highway. Mean classroom concentrations were for all seasons highest in the school along the very busy highway but did not differ much between the two other schools.

The distributions of the personal and home outdoor NO_2 concentrations for children living near highways are presented in Table 6. In all seasons, personal and outdoor NO_2 concentrations were significantly higher for the very busy highway compared to the nonbusy highway. NO_2 concentrations along the fairly busy highway were significantly higher compared to the nonbusy highway in the winter and spring season. For outdoor NO_2 this was also the case in the summer season. Significant differences in personal and outdoor NO_2 concentrations between the very busy and fairly busy highway were found only in the autumn period.

Combining the seasons for 1 year resulted in an estimated difference of $8.2 \mu\text{g}/\text{m}^3$ (SE 1.8) between personal NO_2 exposure of the children from the school with the highest traffic density and the school with the lowest traffic density. Personal NO_2 exposure of the children from the school with the intermediate traffic density was $2.6 \mu\text{g}/\text{m}^3$ (SE 1.4) higher compared to the school with the lowest traffic density. For outdoor NO_2 concentrations these differences were $9.6 \mu\text{g}/\text{m}^3$ (SE 1.1) and 4.9 (SE 0.8), respectively. When total traffic density of the highway as a continuous variable was included in the model, an increase in NO_2 concentrations of $2.6 \mu\text{g}/\text{m}^3$ (SE 0.6) per 50,000 vehicles per 24 hr was estimated for personal exposure and $3.5 \mu\text{g}/\text{m}^3$ (SE 0.4) per 50,000 vehicles per 24 hr for outdoor concentrations. Adding the logarithm of the distance of the home address to the highway to the model did not strongly influence the estimates for traffic density (both categoric and continuous). Outdoor concentrations significantly decreased with increasing distance. The estimated decrease was $-1.3 \mu\text{g}/\text{m}^3$ per $\log(\text{m})$ (SE 0.4). This corresponds to a difference of $1.8 \mu\text{g}/\text{m}^3$ when comparing the concentration at 100-m distance to the concentration at 400-m

Table 4. Distribution of personal and home outdoor NO_2 concentrations of children attending schools in areas with varying degrees of urbanization.

	Personal NO_2 concentrations									Home outdoor NO_2 concentrations								
	Very urban 3,792 addresses/ km^2			Fairly urban 1,481 addresses/ km^2			Nonurban 195 addresses/ km^2			Very urban 3,792 addresses/ km^2			Fairly urban 1,481 addresses/ km^2			Nonurban 195 addresses/ km^2		
	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range
Autumn	14	25.9**	15.3–43.8	47	18.7**	8.6–58.9	41	14.2	7.7–23.8	9	29.8	16.5–36.4	48	27.0**	9.0–34.9	38	24.7	16.9–31.3
Winter	26	42.1**	21.3–163.1	47	23.4**	10.1–45.3	35	19.1	12.5–30.8	20	49.0	30.1–71.4	46	41.5**	25.2–49.5	40	36.1	21.7–43.5
Spring	19	33.3**	13.9–110.4	53	19.6**	4.7–30.1	39	15.8	9.1–23.6	10	38.1**	30.2–70.1	47	29.8**	18.6–36.6	35	22.6	6.1–29.7
Summer	22	19.7**	9.2–30.9	52	17.8**	10.2–29.6	42	14.8	7.2–29.6	14	25.6**	12.9–40.1	51	21.3**	8.3–26.6	37	16.0	7.3–20.8

**Significantly different from the school in the nonurban area; $p < 0.01$.

Table 5. Mean classroom and outdoor school NO_2 concentrations at schools near highways with varying traffic intensities.

Period (dd/mm/yy)	Percentage of time downwind			Classroom mean ^a			Outdoors school		
	Very busy 169,637 v/24 hr	Fairly busy 126,115 v/24 hr	Nonbusy 45,129 v/24 hr	Very busy 169,637 v/24 hr	Fairly busy 126,115 v/24 hr	Nonbusy 45,129 v/24 hr	Very busy 169,637 v/24 hr	Fairly busy 126,115 v/24 hr	Nonbusy 45,129 v/24 hr
Autumn (25/11/97–02/12/97)	66	66	46	19.1	11.9	8.5	— ^b	— ^b	35.1
Winter (09/02/98–16/02/98)	34	34	89	33.5	16.0	19.3	82.3	57.0	46.7
Spring (16/04/98–23/04/98)	53	53	49	25.3	17.1	16.9	36.8	37.1	32.3
Summer (07/07/98–14/07/98)	39	39	67	19.2	12.5	13.0	21.8	17.3	19.5
Average	48	48	63	24.3	14.4	14.4	47.0	37.1	33.4

v, vehicles.

^aAverage of two classrooms. ^bMissing because of vandalism or measurement failure.

Table 6. Distribution of personal and home outdoor NO_2 concentrations of children attending schools near highways with varying traffic intensities.

	Personal NO_2 concentrations									Home outdoor NO_2 concentrations								
	Very busy 169,637 v/24 hr			Fairly busy 126,115 v/24 hr			Nonbusy 45,129 v/24 hr			Very busy 169,637 v/24 hr			Fairly busy 126,115 v/24 hr			Nonbusy 45,129 v/24 hr		
	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range	n	Mean	Range
Autumn	9	26.1*	16.5–37.0	36	19.2	6.4–28.1	41	19.1	9.3–45.7	12	38.0*	26.7–52.9	37	31.9	24.0–37.0	31	34.0	24.2–44.7
Winter	14	36.0*	18.0–82.6	32	29.7*	16.6–46.8	36	25.1	14.1–59.9	20	58.3**	48.5–67.2	35	56.1**	43.1–82.1	33	44.9	36.0–66.9
Spring	15	29.2**	6.0–54.0	32	25.0**	11.9–41.6	37	18.2	6.4–27.3	16	39.3**	10.3–54.7	34	34.9**	26.0–44.8	31	26.9	13.5–35.3
Summer	30	15.6	7.2–25.3	33	15.1	8.9–21.5	31	17.9**	13.7–22.1	33	15.7	9.6–20.1						

*Significantly different from the school along the nonbusy highway; $p < 0.05$. **Significantly different from the school along the nonbusy highway; $p < 0.01$.

distance. For personal NO₂ concentrations the influence of distance of the home address was smaller ($-0.9 \mu\text{g}/\text{m}^3$ per log(m); SE 0.8) and not significant.

Indoor Sources

As expected, having an unvented gas water heater in the home strongly influenced personal NO₂ exposures, with an estimated contribution of $27.0 \mu\text{g}/\text{m}^3$ (SE 4.4) for the urbanization schools and $19.8 \mu\text{g}/\text{m}^3$ (SE 2.7) for the traffic schools. A vented gas water heater had a smaller but also significant influence on personal NO₂ exposures of $5.4 \mu\text{g}/\text{m}^3$ (SE 3.1) and $8.9 \mu\text{g}/\text{m}^3$ (SE 2.5) for the urbanization and traffic schools, respectively. Cooking with gas also significantly contributed to personal NO₂ exposures, with an estimated contribution of $2.4 \mu\text{g}/\text{m}^3$ (SE 0.9) for the urbanization schools and $2.3 \mu\text{g}/\text{m}^3$ (SE 1.3) for the traffic schools. Parental smoking or having a gas space heater in the living room did not significantly influence personal NO₂ exposures.

Differences in personal NO₂ exposures between the urbanization and traffic categories remained significant after adjusting for potential indoor sources. For the urbanization schools, the estimated difference between the very urban and nonurban school, after excluding data from children with either an unvented or vented gas water heater and after adjusting for cooking with gas, parental smoking, and a gas space heater in the living room, was $10.1 \mu\text{g}/\text{m}^3$ (SE 1.2). This is lower than the uncorrected value of $14.6 \mu\text{g}/\text{m}^3$, suggesting that the unadjusted difference was partly caused by indoor sources. The adjusted difference between the fairly urban and nonurban school was $3.3 \mu\text{g}/\text{m}^3$ (SE 0.8), which is similar to the unadjusted value of $3.9 \mu\text{g}/\text{m}^3$. For the traffic schools, the difference between the very busy and nonbusy highway did not change ($8.2 \mu\text{g}/\text{m}^3$; SE 1.6) and the difference between the fairly busy and nonbusy highway increased from $2.6 \mu\text{g}/\text{m}^3$ to $4.5 \mu\text{g}/\text{m}^3$ (SE 1.1). Furthermore, the adjusted model showed a significant decrease in personal NO₂ exposure with increasing distance of the home address from the highway of $-1.4 \mu\text{g}/\text{m}^3$ per log(m) (SE 0.6), whereas in the uncorrected analysis this decrease was smaller and not significant. Including some socioeconomic factors, such as Dutch origin, parental education, or the age of the home, did not change the results.

When all data included in Table 4 were used, the average personal/outdoor ratio was about 0.2 ($p < 0.01$) higher for the very urban school compared to the other two urbanization schools, again pointing toward a stronger influence of indoor sources on personal NO₂ exposures of children from the very urban school. After excluding data from

children with (un)vented gas water heaters in their homes, and after adjusting for cooking with gas, parental smoking, and the presence of a gas space heater in the living room, the difference was less than 0.1 and no longer statistically significant. No significant differences between the personal/outdoor ratios between the traffic schools were found in any of the analyses.

Comparison between Urbanization and Traffic Density Schools

Because measurements were conducted in all six schools simultaneously in autumn, winter, and spring, a direct comparison between schools from the two different parts of the study is possible for these three seasons. Outdoor concentrations at the back side of the schools (Tables 3, 5) were generally higher for the schools along highways compared to the other school with the same degree of urbanization (very busy and fairly busy compared to very urban and not busy compared to fairly urban). Classroom concentrations were, on average, higher in the school along the busiest highway compared to the very urban school and in the school along the least busy highway compared to the fairly urban school. Classroom concentrations, however, were considerably lower in the fairly busy compared to the very urban school. Home outdoor concentrations were significantly higher along highways. The combined season estimated differences were $6.7 \mu\text{g}/\text{m}^3$ and $3.0 \mu\text{g}/\text{m}^3$ for the very busy and fairly busy highway compared to the very urban school, and $2.5 \mu\text{g}/\text{m}^3$ for the nonbusy highway compared to the fairly urban school. Personal exposures, however, were not higher along highways. Personal exposures were actually significantly lower along the fairly busy highway than in the very urban area (estimated difference $5 \mu\text{g}/\text{m}^3$; SE 2).

Discussion

This study has shown that personal and outdoor NO₂ concentrations were significantly different among children living in areas with different degrees of urbanization and among children living in areas close to highways with different traffic densities.

Several other studies have documented significantly higher NO₂ concentrations in urban areas compared to nonurban, suburban, or rural areas (19–22). The estimated differences between the very urban and nonurban area of 10 and $11 \mu\text{g}/\text{m}^3$ for personal and outdoor concentrations, respectively, correlate well with the differences found in other European studies with similar NO₂ levels (19–21). In a study in Helsinki, Finland, personal NO₂ exposures of 246 children 3–6 years of age from eight day-care centers in downtown and suburban areas

measured during 13 weeks were about $9 \mu\text{g}/\text{m}^3$ higher in the downtown area (geometric mean, $26.5 \mu\text{g}/\text{m}^3$) than in the suburban area (geometric mean, $17.5 \mu\text{g}/\text{m}^3$) (19). Krämer et al. (6) measured outdoor and personal NO₂ exposures as part of a study on the health effects of traffic pollution on children living in two urban and one suburban area. Estimated annual personal and outdoor NO₂ concentrations were 5–7 $\mu\text{g}/\text{m}^3$ and 12–17 $\mu\text{g}/\text{m}^3$ higher, respectively, in the urban areas compared to the suburban area.

Less information is available about NO₂ concentrations in city districts along highways with varying traffic densities. The 3 schools that participated in this study were selected out of 24 schools that participated in a study on health effects of exposure to traffic-related air pollution of children attending schools near highways (15). In that study, indoor and outdoor NO₂ concentrations measured at the schools were significantly correlated with total traffic density of the highway. The estimated contribution of total traffic was $3 \mu\text{g}/\text{m}^3$ per 50,000 vehicles for both outdoor and indoor air, which is similar to the values found in this study (2.6 and $3.5 \mu\text{g}/\text{m}^3$ per 50,000 vehicles for personal and home outdoor concentrations, respectively). In a previous study on air pollution near highways in the Netherlands, we also found a significant correlation ($r = 0.68$) between total traffic density and indoor NO₂ concentrations in 12 schools (14). In that study, mean classroom NO₂ concentrations varied between $9.2 \mu\text{g}/\text{m}^3$ at 393 m of a highway with a total traffic density of 81,000 vehicles per 24 hr to $32.8 \mu\text{g}/\text{m}^3$ at 33 m from a highway with 133,000 vehicles per 24 hr. Krämer et al. (6) found a correlation ($r = 0.70$) between outdoor NO₂ and an index characterizing the amount of traffic in front of the child's home. Personal NO₂ exposures were only marginally correlated with outdoor NO₂ ($r = 0.37$).

Personal and home outdoor NO₂ concentrations significantly decreased with increasing distance of the home address to the highway. The estimated decrease was 1.3 and $1.4 \mu\text{g}/\text{m}^3$ per log(m) for home outdoor and personal NO₂, respectively, corresponding to a decrease of about $2 \mu\text{g}/\text{m}^3$ when expressed as the difference between 100 and 400 m distance. Compared to the difference observed between the very busy freeway and the quiet freeway, which was more than $10 \mu\text{g}/\text{m}^3$, this difference was modest, indicating that traffic density on a freeway is a more important determinant of personal NO₂ exposure than distance of home or school from the freeway. For all 24 schools a somewhat higher but nonsignificant value of $1.9 \mu\text{g}/\text{m}^3$ per log(m) was found for both outdoor and classroom concentrations (15). In our previous study on air pollution near highways, we also found a

significant correlation between indoor NO₂ concentrations in classrooms and distance of the school to the highway ($r = -0.83$). Furthermore, outdoor NO₂ measurements at various distances from the same highway showed a clear gradient (14). Two other studies in open terrain downwind of a highway also found a decline in NO₂ concentrations with distance (23,24). Nakai et al. (7) measured personal, indoor, and outdoor NO₂ concentrations in three zones at different distances from two busy roads in Tokyo, Japan. Mean outdoor concentrations decreased from 81 µg/m³ in zone A (< 20 m) to 67 µg/m³ in zone B (20–150 m) and 39 µg/m³ in the reference zone (zone C). Average personal NO₂ exposures in the non-heating season were 60, 56, and 32 µg/m³ for zone A, B, and C, respectively. Two other Japanese studies also documented associations between NO₂ concentrations and distance to major roads (20,25).

Differences between the schools remained significant after excluding children with either vented or unvented gas water heaters in their homes (the two known strongest indoor sources of NO₂ in the Netherlands) and after adjusting for cooking with gas, parental smoking, and a gas space heater in the living room. Including some socioeconomic variables or housing characteristics also did not change the results. In view of our specific research hypothesis, other unidentified indoor sources can only invalidate our conclusions in case the presence of these factors is associated with the degree of urbanization or traffic density. Randomly distributed indoor sources could only have obscured the observed relationship between personal NO₂ and traffic. It is unlikely that any unidentified indoor source is sufficiently strong and sufficiently overrepresented in the highest exposure category to explain the observed differences between the schools. The same holds for other factors that may influence personal NO₂ exposures, such as activity patterns. Furthermore, after adjusting for indoor sources, personal/outdoor ratios did not significantly differ between the schools, which further supports the conclusion that the differences in personal exposures between the schools are most likely caused by differences in outdoor concentrations and not by indoor sources or housing characteristics.

The schools along the very and fairly busy highway were situated in a very urban area, whereas the school along the nonbusy highway was situated in an area with a lower degree of urbanization. As a result, differences between the schools could also have been caused by a difference in urbanization. When comparing the highway schools with the urban schools, home outdoor concentrations were significantly higher for the schools along

highways than for the urbanization school from the area with the same degree of urbanization. No such differences, however, were found for personal NO₂ exposures. Personal NO₂ exposures of children from schools along the fairly busy highway were even significantly lower than for personal exposures of children from the very urban school. Outdoor NO₂ concentrations measured at the back side of these schools were similar. Classroom concentrations, however, were much lower in the school along the fairly busy highway than in the school in the very urban area, especially in the autumn and winter, suggesting a difference in the ventilation of the schools. Nevertheless, personal NO₂ exposures were significantly higher along the very busy highway than those along the fairly busy highway. This difference was also present after taking into account potential indoor sources. As these two schools were in city districts with similar degrees of urbanization, at similar distances from the road, and with the same geographic orientation toward the road, these differences are most likely caused by the difference in traffic densities of the highways. The difference between these two schools and the school along the highway with the lowest traffic density could (partly) have been caused by a difference in urbanization. Conversely, the latter school was situated closer to the road and was about 15% more downwind of the highway than the first two schools during the measurements. This could have resulted in a smaller difference between the fairly busy and nonbusy highway than would have been observed if all schools had been situated at the same distances from the road and had been downwind for a similar percentage of time.

Personal NO₂ exposures were significantly higher for children who lived in homes with a gas-fired water heater, especially when the water heater had no ventilation duct. This is in line with previous Dutch NO₂ monitoring studies, which have shown that these kinds of water heaters are a major source of indoor NO₂ in the Netherlands (17,18). For example, Fischer et al. (18) found an estimated difference in personal NO₂ between women living in homes with and without a gas water heater of 24 µg/m³ in the case of an unvented water heater and 12 µg/m³ in the case of a vented gas water heater. Cooking with gas also significantly increased personal NO₂ concentrations, but this influence was small (± 2 µg/m³) compared to the influence of an unvented gas water heater. This is in line with several other studies that have documented significant higher personal NO₂ concentrations for children (19,26) or adults (22) living in homes where gas is used for cooking. Parental smoking did not significantly influence personal NO₂ exposures. Most of the

studies mentioned previously have found higher personal or indoor NO₂ concentrations for smokers or children exposed to environmental tobacco smoke (19–22,26). The estimated differences, however, are generally small and not always present in all subgroups. For example, in the study among preschool children in Helsinki, personal NO₂ exposures were, on average, about 4 µg/m³ higher for children living with smokers in the suburban area (electric cooking only). In the urban area, however, personal NO₂ exposures were higher (± 3 µg/m³) only for children living with smokers in homes where gas was used for cooking, whereas no difference was found for children living in homes with electric cooking (19).

Conclusion

This study has shown that personal and outdoor NO₂ concentrations are significantly influenced by the degree of urbanization of the city district and by the traffic density of and distance to a nearby highway. As NO₂ can be considered a marker for air pollution from traffic, *a*) degree of urbanization, *b*) traffic density, and *c*) distance to a nearby highway can all be used to estimate exposure to traffic-related air pollution.

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